Met Office

The NAME wet deposition scheme

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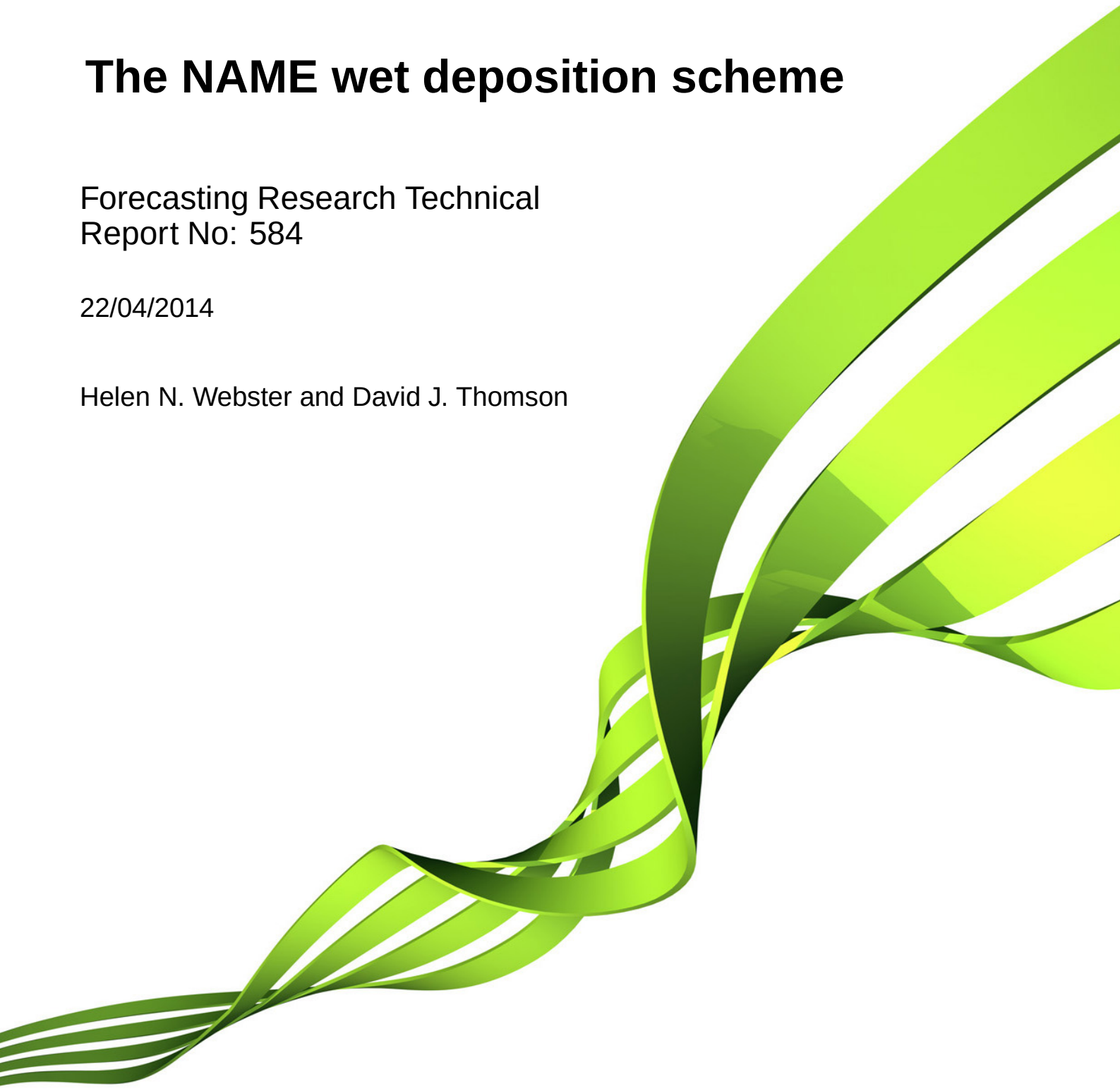

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Abstract

The Numerical Atmospheric-dispersion Modelling Environment (NAME) has had a wet deposition scheme since early on in its development. The original parametrization was developed over 20 years ago and is based on work by Tom Choularton and others at UMIST. Over the years it has largely escaped modifications. Here the original scheme is documented, as far as it possible historically, and reviewed in the context of today's understanding. It is also compared to other parametrizations and assessed for suitability when used with modern NWP meteorological data. As a result, some changes to the original scheme are discussed and implemented.

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1 Wet scavenging

Wet and dry deposition are key mechanisms in the removal of pollutants from the atmosphere. Within regions of precipitation, wet deposition (scavenging by precipitation elements) can be the dominant removal process. Wet deposition depends on the amount, type and intensity of the precipitation and on the properties of the scavenged material, including whether it is in aerosol or gaseous form. For gases, wet deposition depends on the solubility of the gas and gaseous wet scavenging parametrizations are often based on Henry's law. Whereas for aerosols, wet deposition is dependent on the particle size and other particle properties (for example, whether the aerosol is hydrophilic or hydrophobic).

Aerosols within cloud can enter cloud droplets or ice crystals either by the nucleation process (in which the particles act as cloud condensation, or ice, nuclei) or through impaction with the cloud droplets or ice crystals [11]. A fraction of these cloud droplets or ice crystals will then grow into precipitation elements thereby resulting in removal of material from the atmosphere. This process is often referred to as in-cloud scavenging or rainout. Particles in the size range 0.08 μm to around a micrometre are efficient cloud condensing nuclei. Furthermore, nucleation is often the main removal process of sub-micron particles which are usually not efficiently removed by impaction processes. Aerosol mass is primarily scavenged by this method for hydrophilic aerosols. Impaction is an efficient removal process for very small particles with diameters less than 0.01 μm, which undergo Brownian diffusion, and for big particles with diameters larger than about a few micrometres with larger inertial effects.

In addition to impaction between aerosols and cloud droplets / ice crystals in-cloud, aerosols located below the cloud base can be swept out by falling hydrometeors. This wet deposition process is called below-cloud scavenging or washout. It is efficient at removing very small and large particles, for the reasons discussed above in the context of in-cloud scavenging. For particles with diameters between 0.01 μm and a few micrometres, the below-cloud scavenging rate can be up to a few orders of magnitude smaller [14].

Explicit calculation of wet scavenging of aerosols requires consideration of the size distribution and the life cycle of both the aerosols and the cloud droplets or ice crystals. Agglomeration, evaporation, desorption, aggregation and other such processes need to be represented. This is possible in the context of aerosol and cloud microphysics schemes but not usually possible in atmospheric dispersion models which do not have such detailed parametrizations. Bulk parametrizations representing the mean wet scavenging rate for the whole aerosol size range are generally used in atmospheric dispersion models. These bulk schemes assume that the scavenging is equally efficient for aerosols of different sizes. They have been shown, however, to overestimate the scavenging of aerosols by a heavy rain or a long-duration medium rain in comparison to explicit calculations of scavenging [14]. This is due to the fact that, in reality, the mean scavenging coefficient for aerosol mass decreases with time as aerosols in the efficiently scavenged size ranges are removed more

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rapidly from the atmosphere. To address this downfall, Feng [14] designed a wet deposition scheme based on mean scavenging rates for different particle size ranges which could be easily incorporated into traditional atmospheric dispersion models. This scheme therefore partly accounts for the variation in scavenging efficiency of different aerosol sizes without the need for detailed aerosol and cloud microphysics schemes.

Currently, wet deposition is modelled in NAME using a traditional bulk parametrization. The removal of material from the atmosphere by wet deposition processes is based on the depletion equation

$$\frac{\mathrm{d}C}{\mathrm{d}t} = -\Lambda\, C,$$

where C is the air concentration, t is time and Λ is a scavenging coefficient. The mass m of pollutant represented on a NAME model 'particle' is depleted according to

$$\Delta m = m\left[1 - \exp\left(-\Lambda\, \Delta t\right)\right], \tag{1}$$

where Δm is the change in mass over the model time step (Δt). The NAME wet deposition scheme is relatively old and has largely escaped revisions since its development more than twenty years ago. In this paper we review and document the original scheme, as far as we are able, and highlight some particular issues. Some of these issues are due to the passage of time and an incomplete understanding of the scheme in its original form. Other issues have, however, arisen from advances in NWP modelling and computing, since the development of the original NAME wet deposition scheme, and, in particular, with the ability nowadays to conduct high (convective scale) resolution modelling. We describe some modifications implemented to the NAME wet deposition scheme to address these issues.

2 Background to the NAME wet deposition scheme

The NAME wet deposition parametrization dates from around 1991 and is based primarily on work conducted by Tom Choularton and others at UMIST, Manchester, UK. Copies of the original reports from 1991 (including the final report [10]) are stored by the Atmospheric Dispersion and Air Quality group at the Met Office, UK. The suggested parametrizations are based on field studies over moorland at Great Dun Fell and over a forest canopy on Dunslair Heights, on modelling studies and on available literature at the time. The suggested NAME parametrization was intended to be used for wet deposition of radionuclides (the original scope of NAME) although it had been adopted for other species. Most of these other species are aerosols and this seems appropriate since the parametrizations are based on field studies of atmospheric aerosol. However, over the years the original wet deposition parametrization has been adopted for some soluble gaseous species (Ammonia, HNO_3 and HONO) and the reasons for this are not known since different processes are expected to be involved. In contrast, the scheme was also extended to sulphur dioxide, but with

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different scavenging coefficients as expected for a gas with a particular solubility.

3 Occult deposition

We deviate temporarily to discuss occult deposition which is treated, in NAME, as part of the dry deposition scheme. Its origins, however, are based on the wet deposition work of Choularton and Gallagher [10] so, for completeness, it is documented here. Direct turbulent deposition of cloud droplets to the ground generally occurs when a hill-capping cloud forms as moist low level air is forced to ascend over hills. When hill cap clouds form and are in contact with the ground, two physical processes are of relevance: the efficiency with which material is incorporated into the cloud droplets which form during the forced ascent over hills, and factors which affect the deposition of clouds droplets to the ground. These factors depend on the nature of the vegetation, atmospheric turbulence and any surface resistance experienced by the droplets. The deposition velocity is found to be a function of the cloud droplet size, with the surface resistance to deposition evident for droplets with radii less than 5 μm. The forced ascent leads to significant supersaturations at the cloud base and efficient scavenging of soluble material by cloud droplets as they form. The deposition velocities of liquid water have generally been found to be between 90% and 100% of the values for momentum. Taking into account the reduced dry deposition velocity of material not incorporated into the cloud droplets, the dry deposition velocity (v_d) for occult deposition is taken to be 80% of the deposition velocity for heat[1], namely

$$v_d = \frac{0.8}{R_a},$$

where R_a is the aerodynamic resistance.

In NAME, the occult deposition parametrization is invoked if the model particle, representing a certain mass of the pollutant, is less than 50 m above ground level and the dynamic (or total) cloud fraction[2] at the model particle position is greater than 0.5. Dynamic cloud information from the Met Office's Numerical Weather Prediction (NWP) model (the Unified Model (MetUM)) and used by NAME consists of 'low', 'medium' and 'high' cloud amounts but no 'very low' cloud amounts. Currently the MetUM uses a height based η coordinate system. Low, medium and high cloud amounts are defined to be cloud between model levels which fall in the height ranges 111 - 1949 m, 1949 - 5574 m and 5574 - 13608 m, respectively, above sea level from a sea-point grid-box. (The altitudes 111 m, 1949 m, 5574 m and 13608 m equate to pressure levels of 1000 hPa, 800 hPa, 500 hPa and 150 hPa in an ICAO standard atmosphere. Note that the definition of low, medium and high cloud is different in older versions of the MetUM, when a pressure based η coordinate

[1] Choularton and Gallagher [10] refer to the deposition velocity for momentum. Tracer material is thought, however, to be transported in the same way as heat and hence, within NAME, the deposition velocity for heat is used rather than that of momentum

[2] Under the PC2 cloud scheme, currently run in the global configuration of the Met Office's Numerical Weather Prediction model (the Unified Model, MetUM), cloud information is available as total cloud and convective cloud components, whereas prior to the PC2 cloud scheme, cloud information was available as dynamic (large-scale resolved) and convective components. Furthermore, in the high-resolution convection-permitting UKV configuration of the MetUM there is no convection parametrization and hence total cloud is only available.

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system was used, and in other NWP models (e.g., ECMWF).) The dynamic cloud base in NAME is, therefore, constrained with current MetUM data to be no lower than 111 m above sea level for a sea-point grid-box. Consequently there may be situations where the occult deposition scheme is not invoked as it should be. Future consideration should therefore be given to the cloud information taken from the MetUM and, in particular, whether very low cloud amounts should additionally be taken or, alternatively, whether three-dimensional cloud fields should be utilised instead.

4 The original NAME wet deposition parametrization

The scavenging coefficient in the original NAME wet deposition scheme is given by

$$\Lambda = Ar^B, \tag{2}$$

where r is the convective / dynamic precipitation rate (in mm hr^{-1}) and A and B are parameters which varied for different types of precipitation (i.e., dynamic, convective, rain or snow) and for different wet deposition processes (i.e., rainout, washout and the seeder-feeder process). The original parametrization proposal by Choularton [9] and Choularton and Gallagher [10] makes no distinction between convective or dynamic precipitation except for the in-cloud parametrization by snow / ice. In NAME, however, separate wet deposition amounts for dynamic (resolved) precipitation and convective (parametrized) precipitation were calculated in the original scheme and a total wet deposition value obtained by summation of the two components. This raises some interesting issues which are discussed in section 5 (including the incorrect use of the dynamic precipitation wet deposition parametrization with resolved convective precipitation from recent high resolution NWP models).

It has always been acknowledged in NAME that the NWP convective precipitation rate will, in general, underestimate the true convective precipitation rate since it represents the mean rate over the NWP grid-box. In reality, convective showers may be small in comparison to the size of the NWP grid and hence convective precipitation may only be occurring over a fraction of the NWP grid-box. To address this underestimation of convective precipitation rates, the convective precipitation rate (r_{con}) is modified by the convective cloud amount fraction (C_f) to give an effective convective precipitation rate, r_{eff},

$$r_{eff} = \frac{r_{con}}{C_f}.$$

In addition, only a fraction C_f of the particle mass m is depleted according to equation (1).

The total wet deposition was therefore given in NAME by the sum of wet deposition by dynamic and convective precipitation, namely

$$\Delta m = m\left[1 - \exp\left(-\Lambda_{dyn}\,\Delta t\right)\right] + mC_f \exp\left(-\Lambda_{dyn}\,\Delta t\right)\left[1 - \exp\left(-\Lambda_{con}\,\Delta t\right)\right],$$

where Λ_{dyn} and Λ_{con} are dynamic and convective scavenging coefficients, respectively, which de-

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pend on the dynamic and (effective) convective precipitation rates. The mass remaining at the end of the model timestep Δt was therefore given by

$$m(t + \Delta t) = m(t) \left[C_f \exp\left(-\Lambda_{dyn}\, \Delta t\right) \exp\left(-\Lambda_{con}\, \Delta t\right) + \left(1 - C_f\right) \exp\left(-\Lambda_{dyn}\, \Delta t\right) \right]. \quad (3)$$

This formulation is, however, model timestep dependent. That is to say, if $\Delta t = \Delta t_1 + \Delta t_2$ then, ignoring any changes in meteorological variables over Δt, the wet deposition values obtained by using a single model timestep (Δt) will differ from that obtained using two smaller model timesteps (Δt_1 and Δt_2). This arises because the material is (conceptually) removed by convective precipitation from only part of the grid-box but is assumed to be uniformily distributed across the grid-box at the start of the next timestep, meaning that the scheme assumes mixing across the grid-box on a timescale Δt. The timestep, however, may not be related to the actual mixing time. This is clearly unsatisfactory and an issue which we seek to rectify in section 5.1.

4.1 Scavenging coefficients for aerosols

We summarise here the scavenging coefficients, Λ, used in the original NAME wet deposition scheme. For wet scavenging to occur in NAME the (convective or dynamic) precipitation rate must be greater[3] than 0.03 mm hr^{-1}. If the temperature at the NAME particle position is greater than 270 K, then a parametrization for wet deposition by rain was used. Otherwise, a parametrization for wet deposition by snow / ice was adopted. Material above the cloud top is not subject to wet deposition.

4.1.1 The seeder-feeder effect (orographic enhancement)

When cap clouds form over hills, cloud water and dissolved material will be efficiently scavenged by precipitation falling from aloft. This results in an enhancement of the precipitation rate and of the rate of wet deposition of material. The collection efficiencies of raindrops for cloud droplets depend on the raindrop size. For raindrops larger than 0.4 mm, the collection efficiency is 0.9. This decreases to 0.8 for 0.3 mm drops and 0.7 for 0.2 mm drops [10]. In the original NAME wet deposition parametrization a collection efficiency of 0.8 is assumed for raindrops when the feeder cloud is present. The scavenging coefficient (Λ in s^{-1}) for scavenging by rain is given by

$$\Lambda = 4.2 \times 10^{-4}\, E\, r^{0.79},$$

where E is the collection efficiency and r is the total rainfall rate in mm hr^{-1}. Choularton and Gallagher [10] attribute this to Pruppacher and Klett [30] although we have been unable to find

[3] The MetUM has traditionally had a drizzle problem with a tendency to produce too much light precipitation. The use of a critical precipitation value is an attempt to ensure that this over-production of light precipitation does not translate into an over-prediction of wet deposition. Recent work to improve the MetUM drizzle problem may result in removal of this critical precipitation value at some point in the near future.

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reference to this equation in the latest edition. For snowfall, modelling work [10] suggests that the scavenging rate is about three times that for rainfall with the same rain equivalent precipitation rate. Hence, the scavenging coefficient for seeder-feeder scavenging by snow is given by

$$\Lambda = 1.0 \times 10^{-3}\, r^{0.79},$$

where r is the rain equivalent snowfall rate in mm hr^{-1}.

Tom Choularton advises (personal communication) that the seeder-feeder parametrization is applicable to gases and aerosols. Presumably, however, the rate of in-cloud uptake of material into cloud droplets and ice crystals / snowflakes will vary between gases and aerosols and therefore result in different scavenging rates for aerosols and gases by the seeder-feeder process.

Choularton and Gallagher [10] state that these seeder-feeder parametrizations should only be used in the lowest region of the model, since most of the enhancement occurs just above the terrain. They also advise that the precipitation rate should be the surface precipitation rate (i.e., the enhanced rate). The NAME orographic enhancement parametrizations were only intended to be invoked with radar-derived precipitation data when estimates of orographically enhanced precipitation rates are available and an orographic enhancement flag (which used to be available but is not currently) is set, indicating that the seeder-feeder process is taking place. In addition, the orographic enhancement scheme was only applied to material which was less than 1000 m above ground[4], in line with Choularton and Gallagher's recommendation to only use the seeder-feeder parametrization in the lowest region of the model. The seeder-feeder process flag is, however, no longer available as part of the radar data feed and over time the criteria for applying the orographic enhancement parametrization changed. At the time of this review of NAME's wet deposition scheme, the orographic enhancement parametrization was used with all precipitation data (both convective and dynamic) from all sources (both radar and NWP) and was invoked if (i) the model topography (usually taken from the NWP model) exceeded 150 m, and (ii) the model particle (representing the material) was less than 1000 metres above ground level. This is incorrect for two reasons: firstly, the precipitation rate from the NWP model is unlikely (particularly for the lower resolution models) to be representative of the enhanced near surface value, and secondly the orographic enhancement parametrization should only be invoked if the seeder-feeder process is in play and this cannot be determined from topography alone. The underestimation of the near-surface precipitation rate is likely to underestimate the scavenging rate whereas, in contrast, the liberal application of seeder-feeder scavenging coefficients in topographic regions is likely to overestimate the scavenging rate. Furthermore, the on / off nature of the seeder-feeder parametrization at a model topography of 150 m results in an undesirable step change in the wet deposition field (see figure 1). For these reasons, the orographic enhancement scheme has now been removed from NAME until a time when the seeder-feeder process can be inferred from the input data.

[4] NAME documents suggest this may have been 1200 m at some point in the history of NAME

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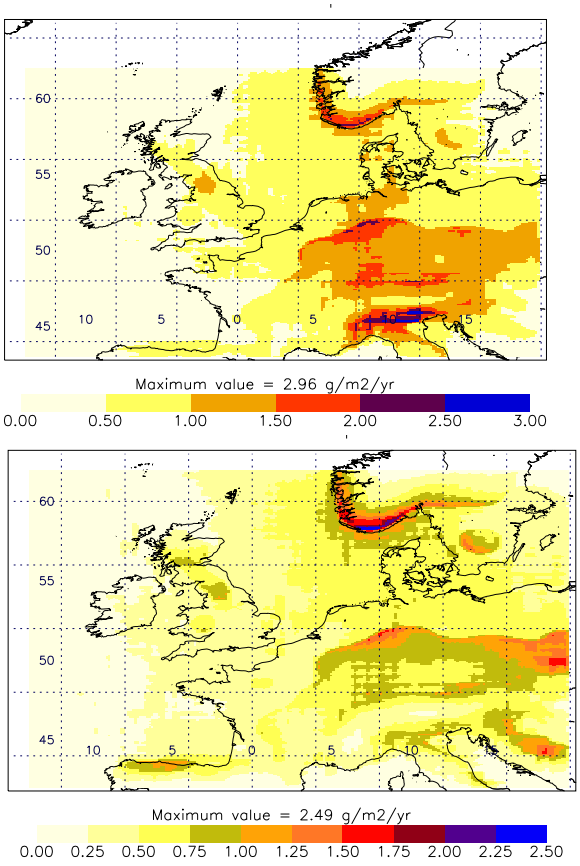

Figure 1: Example annual average wet deposition fields of NO_3^- (upper) and SO_4^{2-} (lower) showing the step change, at an orographic height of 150 m, over continental Europe which is caused by the on / off nature of the seeder-feeder parametrization.

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4.1.2 Below-cloud scavenging

Comprehensive field experiments conducted at Great Dun Fell have been used to deduce a value for the collection efficiency (E) of raindrops for atmospheric aerosol [10]. Data were obtained over the aerosol size range from 0.1 μm to 3.0 μm and results are in good agreement with other measurements published in the literature. For the aerosol diameter range from 0.1 to 1.0 μm, a collection efficiency of between 0.1 and 0.3 is shown to be appropriate. A collection efficiency, E, of 0.2 was chosen to calculate the below-cloud scavenging coefficient, Λ, for rainfall

$$\Lambda = 4.2 \times 10^{-4}\, E\, r^{0.79}.$$

Scavenging of aerosols by snow and ice occurs primarily below-cloud (washout) [22]. Knutson, Sood and Stockham [22] summarise results from field studies. Field studies on scavenging of radioactive material suggest that snow scavenging is more efficient than rain scavenging. However, in contrast, a field study investigating snow scavenging of a tracer aerosol (silver iodide) showed very little difference between rain and snow scavenging. The field studies referred to by Choularton and Gallagher [10] were conducted at high elevation and are in agreement with the results of Knutson and Stockham [23]. The below-cloud scavenging coefficient, Λ, for snowfall was given in the original NAME scheme by

$$\Lambda = 8.0 \times 10^{-5}\, r^{0.305},$$

where r is, as before, the rainfall equivalent precipitation rate in mm hr^{-1} [9]. The r dependence is obtained from field data reported by Knutson and Stockham [23]. According to Choularton and Gallagher [10] this equation for below-cloud scavenging by snow implies a collection efficiency, E, of around 10^{-3} which is consistent with laboratory results of Mitra et al. [27].

The below-cloud parametrizations are adopted if the altitude of the NAME particle is less than the height of the cloud base (convective or dynamic).

4.1.3 In-cloud scavenging

In-cloud scavenging parametrizations apply to particles which lie between the cloud base and the cloud top (convective or dynamic). The dominant precipitation production mechanisms differ between frontal and vigorous convective clouds. This results in different in-cloud parametrizations in the original NAME wet deposition scheme for dynamic and convective precipitation. First we present the main cloud processes involved in the snow and ice phase. These processes, and the in-cloud parametrizations for snow used in the original NAME scheme, are described by Choularton [9].

In convective clouds, supercooled water is present and the dominant growth of cloud ice crystals is via the riming process. This gives rise to precipitation in the form of graupel and hail. The suggested scavenging coefficient, Λ, for convective precipitation in the ice phase within cloud is

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given by

$$\Lambda = 4.2 \times 10^{-4} \, E \, r^{0.79}, \tag{4}$$

where the collection efficiency (E) is taken to be 0.8 and r is the rainfall equivalent precipitation rate in mm hr^{-1} [9].

In dynamic clouds, however, riming is less important and vapour growth of ice crystals followed by aggregation is dominant, resulting in snowfall. In this situation when the cloud is highly glaciated, water droplets will tend to evaporate returning material to the aerosol phase. Under these circumstances, the scavenging rate of the snowflakes for these aerosols will be similar to that which would occur below cloud. Hence the below-cloud parametrization of aerosol scavenging by dynamic snowflakes was used in the original NAME scheme

$$\Lambda = 8.0 \times 10^{-5} \, r^{0.305}. \tag{5}$$

In some dynamic cloud situations, riming of snowflakes, rather than hailstones or graupel (convective cloud situations), can make a contribution. Due to the large surface area of snowflakes, the scavenging rate for snowfall in these conditions is about three times that for rainfall (or hail) with the same rain equivalent precipitation rate. The suggested scavenging coefficient parametrization for such cases [9] is

$$\Lambda = 1.0 \times 10^{-3} \, r^{0.79}. \tag{6}$$

In the original NAME scheme, vapour growth of snowflakes was assumed to be dominant within dynamic cloud. Hence equation (5) was used in preference to equation (6) for the in-cloud scavenging coefficient, Λ.

For the original parametrization of in-cloud scavenging by raindrops, little information exists. In a letter dated from 1995, Tom Choularton states that his thinking on the rainfall rainout parametrization for NAME is that the seeder-feeder parametrization is appropriate. This advice has been followed in the original NAME scheme for raindrops within convective cloud, where the seeder-feeder parametrization with the scavenging coefficient,

$$\Lambda = 4.2 \times 10^{-4} \, E \, r^{0.79}, \tag{7}$$

was used with a collection efficiency, E, of 0.8. For raindrops within dynamic cloud, however, the below-cloud parametrization for the scavenging coefficient

$$\Lambda = 4.2 \times 10^{-4} \, E \, r^{0.79},$$

was used where the collection efficiency was 0.2. The reasons for this different approach and the origins of the original rainout parametrization within dynamic cloud are not known. In fact, one wonders if this was an error in the original implementation.

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4.1.4 Summary of the original NAME wet deposition parametrization

The NAME scavenging parameters (A and B, where the scavenging coefficient $\Lambda = Ar^B$ is in s^{-1} and the precipitation rate r is in mm hr^{-1}) described above and used in the original NAME wet deposition scheme for aerosols are summarised in table 1.

Process	Rain (T $>$ 270 K)		Snow / Ice (T $\leq$ 270 K)	
	Dynamic	Convective	Dynamic	Convective
Orographic enhancement	$A = 3.36 \times 10^{-4}$ $B = 0.79$	$A = 3.36 \times 10^{-4}$ $B = 0.79$	$A = 1.0 \times 10^{-3}$ $B = 0.79$	$A = 1.0 \times 10^{-3}$ $B = 0.79$
Below-cloud	$A = 8.4 \times 10^{-5}$ $B = 0.79$	$A = 8.4 \times 10^{-5}$ $B = 0.79$	$A = 8.0 \times 10^{-5}$ $B = 0.305$	$A = 8.0 \times 10^{-5}$ $B = 0.305$
In-cloud	$A = 8.4 \times 10^{-5}$ $B = 0.79$	$A = 3.36 \times 10^{-4}$ $B = 0.79$	$A = 8.0 \times 10^{-5}$ $B = 0.305$	$A = 3.36 \times 10^{-4}$ $B = 0.79$

Table 1: Scavenging parameters for aerosols used in the original NAME wet deposition scheme

4.2 Scavenging coefficients for gases

Most gases are relatively insoluble and hence wet deposition of gases is generally neglected in NAME. The NAME aerosol wet deposition scheme described in section 4.1 was, however, oddly applied for the following soluble gases: ammonia (NH_3), nitric acid (HNO_3) and nitrous acid (HONO). This issue is discussed further in section 5. For sulphur dioxide (SO_2), a simple gaseous scavenging scheme was employed with fixed scavenging parameters $A = 2.4 \times 10^{-5}$ and $B = 0.79$ used in all situations (in-cloud / below-cloud, convective precipitation / dynamic precipitation, rain / snow or ice). In NAME II (the predecessor to NAME III) there is a comment in the Fortran code stating that A was taken from the ratio of the EMEP scavenging coefficients for SO_4 (sulphate - an aerosol) and SO_2. The same dependence of the scavenging rate on precipitation as is used for aerosols (i.e., through the B scavenging parameter) is maintained (in most situations). The ratio of the SO_2 and SO_4 EMEP scavenging coefficients, $\Lambda_{SO2}/\Lambda_{SO4}$, is 0.3 in-cloud and 1.44 below-cloud (see appendix B.6 and table 11). Maintaining the EMEP ratio between the NAME scavenging coefficients for SO_2 and SO_4 (for which the NAME scavenging parameters for aerosols given in table 1 are used) gives the SO_2 scavenging parameters in table 2. The dynamic in-cloud values for rain in table 2 are roughly those originally chosen for SO_2 scavenging in NAME but it may be worth considering variations for rain / snow, in-cloud / below-cloud, etc. along the lines of table 2, although it is not clear that scavenging of SO_2 varies with snow / rain etc. in the same way as scavenging of SO_4. Furthermore, different scavenging parameters should perhaps be chosen for other soluble gases (e.g., ammonia (NH_3), nitric acid (HNO_3) and nitrous acid (HONO)) rather than using aerosol scavenging parameters as was the case in the original scheme. This is discussed further in section 5.2 and suggested revised

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scavenging parameters for gases are given in table 4.

Process	Rain (T $>$ 270 K)		Snow / Ice (T $\leq$ 270 K)	
	Dynamic	Convective	Dynamic	Convective
Below-cloud	$A = 1.2 \times 10^{-4}$ $B = 0.79$	$A = 1.2 \times 10^{-4}$ $B = 0.79$	$A = 1.2 \times 10^{-4}$ $B = 0.305$	$A = 1.2 \times 10^{-4}$ $B = 0.305$
In-cloud	$A = 2.5 \times 10^{-5}$ $B = 0.79$	$A = 1.0 \times 10^{-4}$ $B = 0.79$	$A = 2.4 \times 10^{-5}$ $B = 0.305$	$A = 1.0 \times 10^{-4}$ $B = 0.79$

Table 2: Possible SO_2 scavenging parameters for NAME, maintaining the EMEP ratio between the SO_2 and SO_4 (aerosol) scavenging coefficients.

5 A discussion of some issues with, and subsequent modifications to, the original NAME wet deposition scheme

In section 4.1.1 we stated that the orographic enhancement scheme, which was being incorrectly applied, has been removed until such time that the seeder-feeder process can be determined from the input meteorological data. The user should note, however, that this decision is likely to result in a reduction in the wet deposition predicted by NAME in orographic regions. Furthermore, NAME may underpredict wet deposition when the seeder-feeder process is, in reality, occurring since the enhanced level of wet scavenging (due to an enhanced collection efficiency) is now not being represented.

In this section we discuss some other issues concerning the original NAME scheme and describe recent revisions made to the wet deposition scheme to address these issues. These changes to the wet deposition parametrization have been implemented in version 6.3 of NAME.

5.1 Dynamic (resolved) and convective (parametrized) precipitation

In the original NAME scheme, wet deposition comprised of two elements: scavenging by dynamic precipitation and scavenging by convective precipitation. Total wet deposition was obtained by summing the convective and dynamic components.

The idea behind this split into dynamic and convective components was probably partly influenced by the differences in scales of the precipitation features, with convective showers potentially much smaller than the spatial resolution of the NWP model. The true precipitation rate within convective showers will be greater than the NWP model grid-box mean precipitation rate and, furthermore, the spatial extent of the convective shower will be less than that implied by the NWP model. In calculating the convective component of wet deposition NAME therefore modifies the convective precipitation rate, and the amount of material subject to wet deposition, by the convective cloud

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amount.

The division between convective and dynamic precipitation in the original NAME wet deposition scheme is, however, also meant to represent the different physical processes at play in stratiform and convective clouds. The original documents by Choularton [9] and Choularton and Gallagher [10], however, do not generally distinguish between large-scale / dynamic or convective precipitation except in the case of in-cloud scavenging by frozen precipitation.

Large-scale or dynamic precipitation was taken in NAME to be the precipitation resolved by the NWP model whereas convective precipitation was assumed to be precipitation from the convection parametrization. In recent years, however, the division in NWP models between 'dynamic' and 'convective' (or resolved and parametrized) precipitation has blurred as increasing resolution of NWP models means that convective precipitation has begun to be resolved. Indeed, the high resolution UKV configuration of the MetUM (1.5 km horizontal resolution in the interior) does not run a convection scheme with all precipitation resolved by the model. Hence the resolved precipitation from the UKV configuration of the MetUM represents total precipitation not just the large-scale / dynamic (non-convective) component as previously assumed. This raises a few issues with the NAME wet deposition scheme in its original form.

Since in the NAME parametrization the scavenging coefficient (Λ in equation (2)) is nonlinear in the precipitation rate ($B \neq 1$), the predicted wet deposition depends on the ratio of convective (parametrized) to dynamic (resolved) precipitation, even if the dynamic and convective scavenging coefficients are the same and the convection cloud fraction is 1.0. As discussed above, this ratio is likely to vary with NWP model resolution. When the same A and B scavenging parameters were used for dynamic and convective precipitation (i.e., all cases in the original parametrization except in-cloud, see table 1), the difference between the wet deposition based on total precipitation and the sum of the convective and dynamic wet deposition components is likely to be small either when B is close to one (i.e., when the scavenging coefficients are near-linear in precipitation) or when the precipitation is dominated by either convective (parametrized) or dynamic (resolved) precipitation.

In the original in-cloud parametrizations the A and B parameters differed between dynamic and convective precipitation (table 1). With the higher resolution NWP models currently available, the dynamic in-cloud scavenging parameters could be incorrectly employed with convective precipitation when convection is resolved. To prevent this happening, we choose to modify the original in-cloud NAME scheme so that equivalent A and B parameters are used for both dynamic (resolved) and convective (parametrized) precipitation.

For in-cloud scavenging of aerosols by rain, we select the original convective parametrization since the origin of the in-cloud parametrization by dynamic rain is not traceable in detail. This is also in close agreement with the in-cloud parametrization used by Feng (personal communication) where $\Lambda = RAr^B$, $A = 5.2 \times 10^{-4}$ s^{-1}, $B = 0.8$ and R is the in-cloud scavenging ratio[5] as given in table 3 of Stier et al. [45] or table 1 of Croft et al. [11]. Based on the assumption that aerosol

[5]The scavenging ratio is defined as the fraction of the aerosol in the cloud that is embedded in the cloud liquid water / ice.

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mass mainly lies within the accumulation mode (particle radii from 0.05 μm to 0.5 μm), $R = 0.85$ for soluble aerosols in liquid stratiform clouds [11, 45] and hence $\Lambda = 4.4 \times 10^{-4} r^{0.8}$ which is similar to the original NAME in-cloud scavenging rate by convective rain.

For in-cloud scavenging by ice, nucleation[6] of aerosols in ice clouds is not very efficient. The in-cloud scavenging ratio (R) in stratiform ice clouds given in table 3 of Stier et al. [45] and in table 1 of Croft et al. [11] is consequently small ($R = 0.1$) for all aerosol sizes and types. Hence, Feng's in-cloud parametrization for ice stratiform clouds (personal communication) has a scavenging coefficient given by $\Lambda = 5.2 \times 10^{-5} r^{0.8}$. NAME's in-cloud 'A' scavenging parameter for dynamic ice / snow is similar to Feng's 'RA' coefficient. However, the 'B' parameters differ significantly with the low 'B' value ($B = 0.305$) used in the original NAME in-cloud parametrization for dynamic snow / ice significantly below those 'B' values used in other models (see appendix B). (Note this also applies to the below-cloud 'B' scavenging parameter for snow / ice and hence this may be investigated at a later date.) The scavenging parameters $A = 5.2 \times 10^{-5}$ s^{-1} and $B = 0.79$ are therefore adopted in NAME for in-cloud scavenging by snow or ice, in line with Feng's values.

We note, however, that the in-cloud scavenging ratios given in table 1 of Croft et al. [11] and in table 3 of Stier et al. [45] are based on the following definitions of liquid, mixed phase and ice clouds: liquid (T $>$ 273.15 K), mixed phase (238.15 $<$ T $\leq$ 273.15 K) and ice (T $\leq$ 238.15 K). Hence we modify NAME to use the ice / snow scavenging parameters in-cloud when T $\leq$ 238.15 K and to use the rain scavenging parameters in-cloud when T $>$ 273.15 K. At temperatures between (238.15 $<$ T $\leq$ 273.15 K) we choose to linearly interpolate the in-cloud 'A' scavenging parameter between the rain and ice values ($B = 0.79$ in-cloud throughout) in the belief that the amount of supercooled liquid within the cloud reduces with decreasing temperature. We note, however, that some consideration could be given in the future to using the in-cloud scavenging ratios (R) for mixed phase stratiform clouds given in table 1 of Croft et al. [11] and in table 3 of Stier et al. [45] although this may give a more significant change to the wet deposition amounts predicted by NAME than the approach adopted here (and a step change at temperatures of 238.15 K and 273.15 K). In addition, some consideration should also be given to adopt the in-cloud scavenging ratios for insoluble aerosols (instead of those for soluble aerosols) for hydrophobic aerosols such as dust.

Finally, we mention that the in-cloud scavenging ratios of Croft et al. [11] and Stier et al. [45] used here are for stratiform clouds. Stier et al. [45] state that there are few size-resolved measurement data for convective clouds. For mixed phase convective clouds, the in-cloud scavenging ratios given by Stier et al. [45] are based on increasing the values used for mixed phase stratiform clouds, in the aerosol size ranges of potential cloud condensing nuclei. This assumes higher supersaturations in convective clouds. It is not clear what values are recommended for in-cloud scavenging ratios for liquid and ice convective clouds. Since they are not mentioned explicitly by Stier et al. [45], either the same values as for liquid and ice stratiform clouds may be intended or precipitating convective clouds may be regarded as being generally of mixed phase. Either way, since the revisions to

[6] Nucleation is often the main in-cloud removal process

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the NAME wet deposition scheme have removed the distinction between convective and dynamic clouds, we adopt Stier et al.'s [45] stratiform values. Table 3 summarises these modified scavenging parameters recommended for use in NAME. Note that the temperature which determines if the rain or snow / ice parametrization should be used below cloud remains unchanged at T = 270 K.

To enable more flexibility and to ease tuning of the wet deposition scheme by the user, the 'A' and 'B' scavenging parameters have been added to the species block in the NAME input file. There are eight relevant column keywords under the species headed block: 'A rain - BC', 'B rain - BC', 'A snow - BC', 'B snow - BC', 'A rain - IC', 'B rain - IC', 'A snow - IC' and 'B snow - IC', where 'BC' stands for 'below-cloud' and 'IC' stands for 'in-cloud'. Negative values are not permitted and leaving all of these eight values blank results in no wet deposition. The column keyword 'Wet Dep?' is now superfluous and has been removed from the species headed block.

Process	Rain	Snow / Ice
Below-cloud	**T $>$ 270 K** $A = 8.4 \times 10^{-5}$ $B = 0.79$	**T $\leq$ 270 K** $A = 8.0 \times 10^{-5}$ $B = 0.305$
In-cloud	**T $>$ 273.15 K** $A = 3.36 \times 10^{-4}$ $B = 0.79$	**T $<$ 238.15 K** $A = 5.2 \times 10^{-5}$ $B = 0.79$

Table 3: Suggested revised scavenging parameters for aerosols in NAME. The in-cloud parametrization also has a mixed phase (for temperatures between 238.15 K and 273.15 K) where A and B are linearly interpolated between the snow / ice and rain values.

We return to the issue of the dependency of NAME predicted wet deposition on the ratio of the parametrized to resolved precipitation. The original scheme in NAME calculated total wet deposition by summing the individual convective and dynamic wet deposition components. In the revised scheme, with equivalent 'A' and 'B' scavenging parameters used for 'convective' and 'dynamic' precipitation, an overall total scavenging coefficient Λ_{tot} can be calculated by splitting the MetUM model grid-box into two parts: a fraction C_f, where C_f is the convective cloud fraction, which is within the region of convective precipitation and where the total precipitation rate is given by $r_{dyn} + r_{con}/C_f$, and the remaining fraction $(1 - C_f)$ outside of the region of convective precipitation where the total precipitation rate is given by r_{dyn}. The overall total scavenging coefficient is given by

$$\Lambda_{tot} = (1 - C_f)A_1 r_{dyn}^{B_1} + C_f A_2 \left(r_{dyn} + \frac{r_{con}}{C_f} \right)^{B_2}, \tag{8}$$

where the appropriate in-cloud / below-cloud and rain / snow scavenging coefficients are chosen for A_1, A_2, B_1 and B_2. The in-cloud values are used for A_1 and B_1 if the NAME particle lies between the dynamic (resolved) cloud base and the dynamic cloud top[7] and the below-cloud values

[7] Under the PC2 cloud scheme (currently run in the global MetUM) dynamic cloud information is not available and hence total cloud is used in its place here

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are used if the NAME particle lies below the dynamic (resolved) cloud base. The in-cloud values are used for A_2 and B_2 if the NAME particle lies between Min(convective cloud base, dynamic cloud base) and Max(convective cloud top, dynamic cloud top) whereas the below-cloud values are used if the NAME particle lies below Min(convective cloud base, dynamic cloud base). Equation (8) also has the advantage of being model timestep independent and gives the correct limits for purely convective ($\Lambda = C_f A(r_{con}/C_f)^B$) or purely dynamic ($\Lambda = A r_{dyn}^B$) precipitation. It also gives the correct limit for increasing NWP model resolution when $C_f \rightarrow 0$ and r_{dyn} becomes the total precipitation at convection resolving scales. The dependency of NAME predicted wet deposition on the ratio of parametrized to resolved precipitation has been removed by equation (8) for the case when $C_f = 1$. NAME has now been revised to use equation (8) instead of calculating individual convective and dynamic wet deposition components.

5.2 Gases

As indicated earlier, specific scavenging parameters, based on solubility, are recommended for soluble gases: SO_2, H_2S, NH_3, HNO_3, H_2O_2 and HONO. For SO_2, the EMEP scavenging ratio between SO_4 and SO_2 is maintained between the NAME scavenging coefficients for aerosols and SO_2. H_2S is less soluble than SO_2 with the belief that wet deposition of H_2S is between 2 and 10 times less than wet deposition of SO_2. A mid-value of 6 times less is chosen here. For NH_3 and HNO_3, 'A' scavenging parameters within the range of values used by other models and given in the literature (see appendices A and B) are chosen. Scavenging parameters for HNO_3 are also adopted for HONO and H_2O_2. These recommended scavenging parameters for gases (see table 4) are first approximations and may need to be tuned further. In particular, the (below-cloud) scavenging coefficient suggested for SO_2 seems high in comparison to those for NH_3 and HNO_3, based on other models and values given in the literature. Furthermore, the low 'B' scavenging parameter, which NAME still uses for below-cloud scavenging by snow and ice, may need some investigation as it appears to be unsupported by other schemes / models.

6 Sensitivity to the scavenging coefficients

Despite only subtle changes to the scavenging coefficients in the NAME wet deposition scheme, the effect on predicted wet deposition and air concentration fields can be significant, indicating a high sensitivity to the scavenging coefficients. As an example we show here a volcanic ash run which is particularly sensitive to the in-cloud scavenging by snow / ice parametrization. Figure 2 shows the predicted six-hourly averaged air concentrations of volcanic ash between FL000 and FL200, where FL are flight levels which are roughly equivalent to hundreds of feet, obtained using the original in-cloud and below-cloud scavenging coefficients as described in table 1. Note that the original method of calculating total wet deposition by summing convective and dynamic components

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Species	Below-cloud		In-cloud†	
	rain (T $>$ 270 K)	snow / ice (T $\leq$ 270 K)	rain (T $>$ 273.15 K)	snow / ice (T $<$ 238.15 K)
Aerosols	$A = 8.4 \times 10^{-5}$ $B = 0.79$	$A = 8.0 \times 10^{-5}$ $B = 0.305$	$A = 3.36 \times 10^{-4}$ $B = 0.79$	$A = 5.2 \times 10^{-5}$ $B = 0.79$
SO_2	$A = 1.2 \times 10^{-4}$ $B = 0.79$	$A = 1.2 \times 10^{-4}$ $B = 0.305$	$A = 1.0 \times 10^{-4}$ $B = 0.79$	$A = 1.5 \times 10^{-5}$ $B = 0.79$
Ammonia	$A = 1.2 \times 10^{-4}$ $B = 0.79$	$A = 1.2 \times 10^{-4}$ $B = 0.305$	$A = 3.5 \times 10^{-4}$ $B = 0.79$	$A = 3.5 \times 10^{-4}$ $B = 0.79$
HNO_3, H_2O_2 & HONO	$A = 1.0 \times 10^{-4}$ $B = 0.79$	$A = 1.0 \times 10^{-4}$ $B = 0.305$	$A = 3.5 \times 10^{-4}$ $B = 0.79$	$A = 3.5 \times 10^{-4}$ $B = 0.79$
H_2S	$A = 2.0 \times 10^{-5}$ $B = 0.79$	$A = 2.0 \times 10^{-5}$ $B = 0.305$	$A = 1.7 \times 10^{-5}$ $B = 0.79$	$A = 2.6 \times 10^{-6}$ $B = 0.79$

Table 4: Suggested NAME scavenging parameters for different species. (†In the mixed phase (238.15 K $<$ T $<$ 273.15 K), the in-cloud 'A' and 'B' scavenging parameters are given by linearly interpolating between the rain and snow / ice values)

is used here but the orographic enhancement scheme has been removed.

With the removal of the convective / dynamic split we tested different parametrizations for in-cloud scavenging. The effect of these different in-cloud scavenging coefficients on predicted six-hourly averaged air concentrations for this particular volcanic ash example are shown in figures 3 to 6. Note that an overall total scavenging coefficient as given by equation (8) is calculated and the original below-cloud scavenging parameters (see table 1) are used throughout. Table 5 summarises the differences between the in-cloud parametrizations used in figures 3 to 6. The original in-cloud scavenging parameters for convective rain ($A = 3.36 \times 10^{-4}$, $B = 0.79$) are used for the in-cloud rain parametrization.

Figure 3 shows the NAME predicted ash cloud between FL000 and FL200 using the original convective scavenging parameters in-cloud with both resolved and parametrized precipitation. The, in general, larger in-cloud scavenging parameters (particularly for snow / ice) result in significantly more wet deposition and hence lower ash concentrations within the volcanic plume.

In figure 4 the original dynamic scavenging parameters have been used in-cloud with resolved and parametrized snow / ice. (The original convective in-cloud scavenging parameters have been used with rain.) This results, in general, in a slightly smaller scavenging coefficient and hence slightly higher air concentrations than the original scheme shown in figure 2.

Based on discussions with Feng (personal communication) concerning in-cloud scavenging rates by snow / ice and the work or Stier et al. [45], the scavenging parameters $A = 5.2 \times 10^{-5}$ and $B = 0.79$ for in-cloud scavenging by snow / ice were tested in NAME. (Note that the original convective scavenging parameters are again used in-cloud with rain.) The original temperature definitions for rain and snow / ice (T $>$ 270 K and T $\leq$ 270 K, respectively) were used. Figure 5

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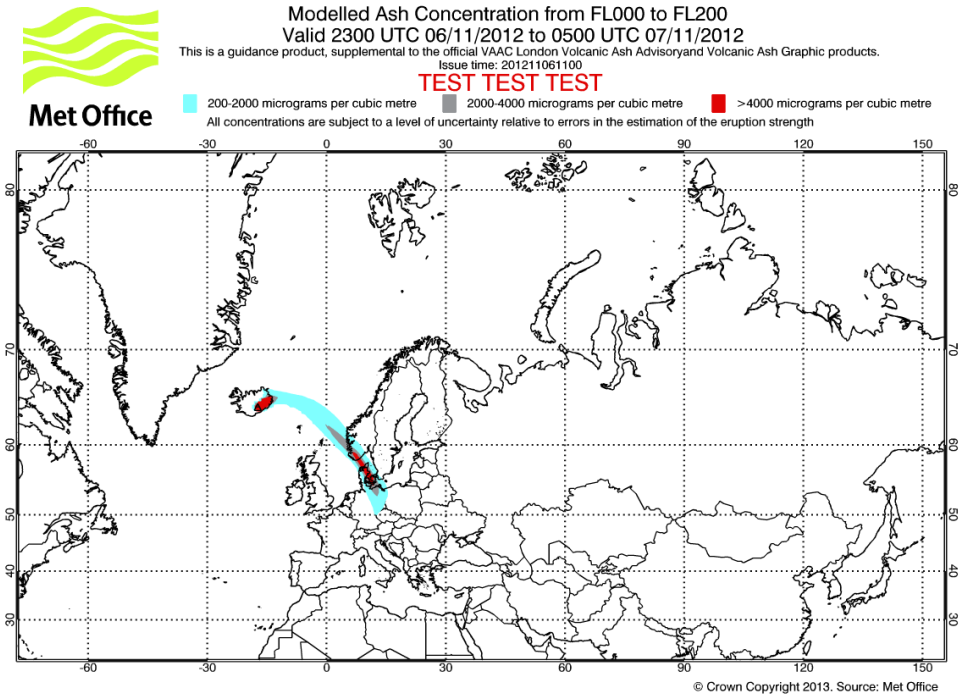

Figure 2: Example air concentration field from a volcanic ash run using the original NAME wet deposition scheme in-cloud and below-cloud as described in table 1

Figure	In-cloud scavenging parameters		Further information
	rain	snow / ice	
3	$A = 3.36 \times 10^{-4}$ $B = 0.79$	$A = 3.36 \times 10^{-4}$ $B = 0.79$	rain: T $>$ 270 K snow: T $\leq$ 270 K
4	$A = 3.36 \times 10^{-4}$ $B = 0.79$	$A = 8.0 \times 10^{-5}$ $B = 0.305$	rain: T $>$ 270 K snow: T $\leq$ 270 K
5	$A = 3.36 \times 10^{-4}$ $B = 0.79$	$A = 5.2 \times 10^{-5}$ $B = 0.79$	rain: T $>$ 270 K snow: T $\leq$ 270 K
6	$A = 3.36 \times 10^{-4}$ $B = 0.79$	$A = 5.2 \times 10^{-5}$ $B = 0.79$	rain: T $>$ 273.15 K snow: T $\leq$ 238.15 K‡

Table 5: A summary of the differences between the in-cloud parametrizations tested in Figures 3 to 6. ‡In the mixed phase regime ($238.15 \leq T \leq 273.15$ K) the 'A' and 'B' scavenging parameters are linearly interpolated between the rain and snow / ice values.

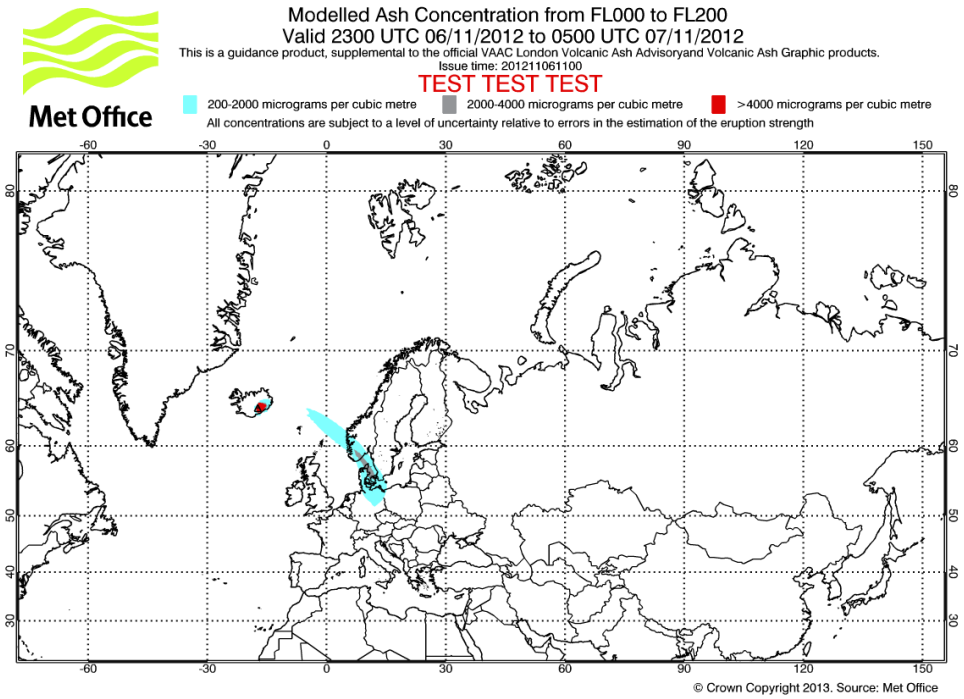

Figure 3: Example air concentration field from a volcanic ash run using the original convective scavenging parameters ($A = 3.36 \times 10^{-4}$, $B = 0.79$) for rain and snow

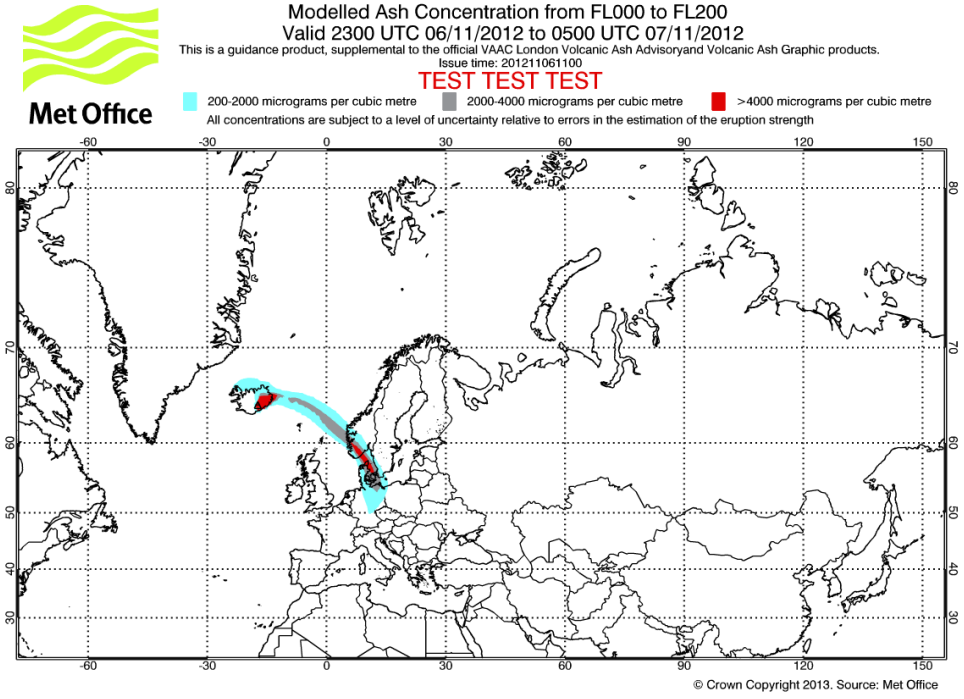

Figure 4: Example air concentration field from a volcanic ash run using the original convective in-cloud scavenging parameters with rain ($A = 3.36 \times 10^{-4}$, $B = 0.79$) and the original dynamic in-cloud scavenging parameters with snow / ice ($A = 8.0 \times 10^{-5}$, $B = 0.305$).

shows the volcanic ash concentrations using these in-cloud scavenging parameters. The in-cloud scavenging rate is much smaller and hence considerably higher air concentrations are predicted than with the original scheme (cf. figure 2).

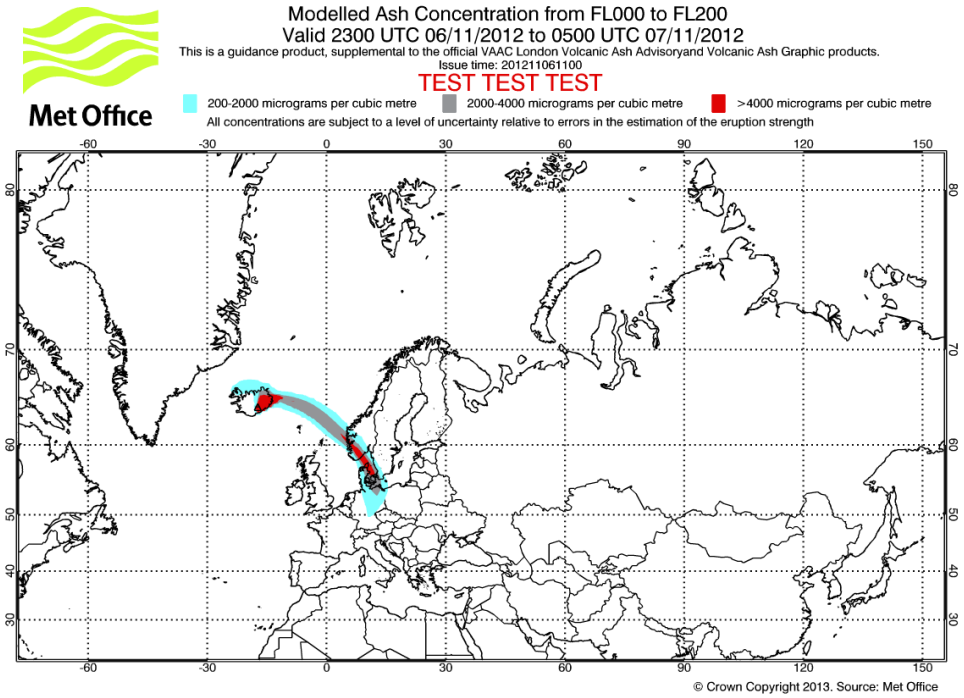

Figure 5: Example air concentration field from a volcanic ash run using the original convective in-cloud scavenging parameters with rain ($A = 3.36 \times 10^{-4}$, $B = 0.79$) and the scavenging parameters $A = 5.2 \times 10^{-5}$, $B = 0.79$ in-cloud with snow / ice

Stier et al. [45] have different definitions of rain (T $>$ 273.15 K) and snow / ice (T $<$ 238.15 K) than the original NAME wet deposition scheme. In addition, they have a mixed phase for temperatures in between (238.15 $<$ T $<$ 273.15 K). Figure 6 shows the predicted concentration obtained using the same in-cloud rain and snow / ice scavenging coefficients as figure 5 but with Stier's definition of rain or snow / ice used in-cloud. In the mixed phase region the 'A' and 'B' scavenging parameters are obtained by linearly interpolating between the rain and snow / ice values. Note that the scavenging parameters and the definitions of rain, snow / ice and mixed phase precipitation used in figure 6 are those employed in the revised scheme in version 6.3 of NAME. Figure 6 shows that there is a slight increase in wet deposition and consequently a slight decrease in predicted air concentrations over the original NAME wet deposition scheme (see figure 2). However, the differences are not as stark as some of the other options tested here.

7 Summary

The original NAME wet deposition parametrization, derived by Tom Choularton, dates from around 1991 and was based on data from field studies, on modelling studies and on values in the literature

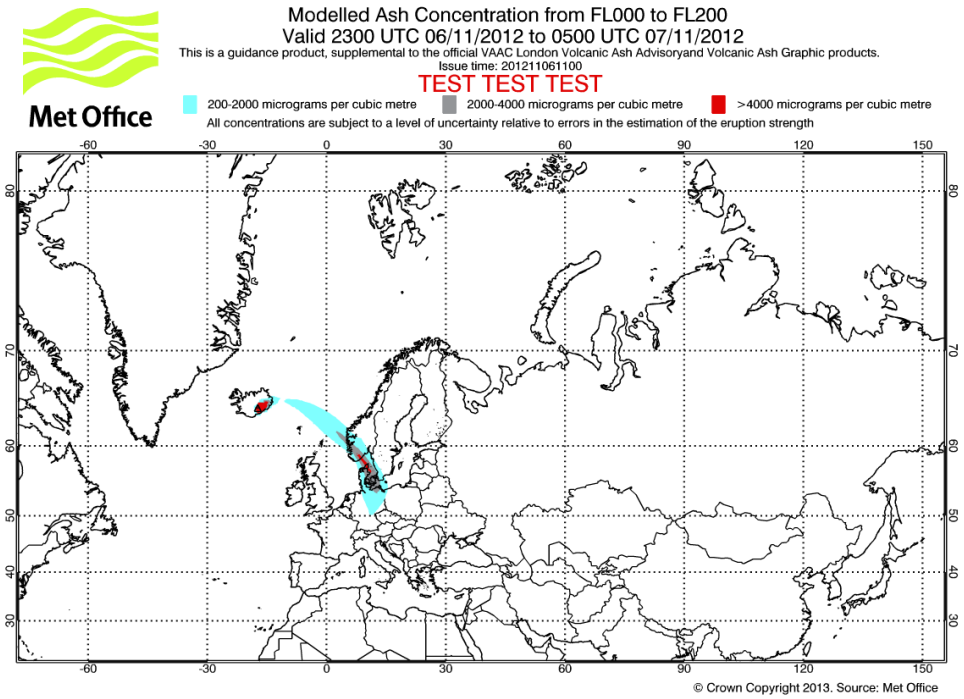

Figure 6: Example air concentration field from a volcanic ash run using the original convective in-cloud scavenging parameters ($A = 3.36 \times 10^{-4}$, $B = 0.79$) with rain (T $>$ 273.15 K) and the scavenging parameters $A = 5.2 \times 10^{-5}$, $B = 0.79$ with snow / ice (T $<$ 238.15 K), with interpolated values in the mixed phase (238.15 $<$ T $<$ 273.15 K)

available at the time. Its original scope was for modelling wet deposition of radionuclides but it is also applicable for wet deposition of aerosols. It is not, however, applicable for wet deposition of soluble gases and different scavenging coefficients have now been implemented for these, based on scavenging coefficients used in other models (see appendix B) and given in the literature (see appendix A).

There are some issues concerning the implementation of the Choularton parametrization in NAME. Firstly, there is very little information on the 'in-cloud scavenging by rain' parametrization originally used in NAME and consequently the origin of this scheme is not fully understood. Choularton does not appear to distinguish between in-cloud scavenging by convective and large-scale rain, whereas the original NAME parametrization does. Secondly, it is clear that the seeder-feeder (or orographic enhancement) parametrization was only intended to be used when the seeder-feeder process is in play. It is not obvious how this process could currently be determined from the available input meteorological data. Lastly, in the NAME implementation of the Choularton scheme, the wet deposition amounts from convective (parametrized) and dynamic (resolved) precipitation were calculated separately, even where there is no distinction in the scavenging coefficients between convective and large-scale precipitation. Because of the non-linearity of the parametrization, summing the convective and dynamic wet deposition components to obtain the total wet deposition will give different results to those obtained from a total precipitation value given by adding the convective and large-scale precipitation rates.

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Increases in NWP model resolution have, however, changed the division between model resolved and model parametrized precipitation, with the recent UKV configuration of the MetUM in the extreme situation of having all resolved, and no parametrized, precipitation. The original NAME in-cloud wet deposition parametrization differed for resolved and parametrized precipitation, acknowledging that different processes are involved in convective and dynamic clouds (for example, different processes are involved in the formation of snow and ice particles). However, this meant that, when convection is resolved in the higher resolution models, NAME was incorrectly using the dynamic precipitation parametrization with convective precipitation. For this reason the scheme has been modified to use equivalent scavenging parameters for dynamic and convective precipitation. This allows a total scavenging coefficient to be calculated from dynamic and convective components. The resulting parametrization is time-step independent and the total scavenging coefficient is not, for a convective cloud fraction of 1, dependent on the ratio of NWP model resolved to NWP model parametrized precipitation and has the appropriate limits in totally convective or totally dynamic situations. It is acknowledged that the user may wish to change the scavenging parameters used by NAME and, for this reason, they are no longer hardwired in the model and have been moved into the species headed block in the input file for ease of modification. Table 6 summarises the wet deposition column headers in the Species headed block for version 6.3 of NAME and the suggested values for different species.

Column header	Aerosols	SO_2	Ammonia	HNO_3, H_2O_2 & HONO	H_2S
A rain - BC	8.4×10^{-5}	1.2×10^{-4}	1.2×10^{-4}	1.0×10^{-4}	2.0×10^{-5}
B rain - BC	0.79	0.79	0.79	0.79	0.79
A snow - BC	8.0×10^{-5}	1.2×10^{-4}	1.2×10^{-4}	1.0×10^{-4}	2.0×10^{-5}
B snow - BC	0.305	0.305	0.305	0.305	0.305
A rain - IC	3.36×10^{-4}	1.0×10^{-4}	3.5×10^{-4}	3.5×10^{-4}	1.7×10^{-5}
B rain - IC	0.79	0.79	0.79	0.79	0.79
A snow - IC	5.2×10^{-5}	1.5×10^{-5}	3.5×10^{-4}	3.5×10^{-4}	2.6×10^{-6}
B snow - IC	0.79	0.79	0.79	0.79	0.79

Table 6: Wet deposition column headers in the species block for version 6.3 of NAME and suggested values for the scavenging parameters. Note that BC denotes below-cloud and IC denotes in-cloud.

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A Wet scavenging coefficients in the literature

Pruppacher and Klett [30] note that scavenging coefficients for aerosols generally range between 1×10^{-5} and 1×10^{-3} s^{-1}. These values are consistent with data of Huebert et al. [17], Schumann [32], Schumann et al. [33] and Jylhä [20]. Scavenging coefficients for SO_2 have been obtained by Beilke [4] from laboratory data and by Makhon'ko [25], Hales et al. [15] and Dana et al. [12] from field observations. Typical scavenging coefficients for SO_2 range between 5×10^{-6} and 6×10^{-5} s^{-1}. Field measurements for the more soluble gases, NH_3 and HNO_3, are given by Huebert et al. [17] and by Shimshock and de Pena [38]. Typical scavenging coefficients for these gases range between 1×10^{-4} and 3×10^{-4} s^{-1}.

A.1 Wet deposition parametrizations of aerosols

Wet deposition of aerosols has a strong sensitivity to both the particle and raindrop sizes. However, simple bulk parametrizations, such as that used in NAME, which depend only on the precipitation rate and not on the particle size and the raindrop distribution, are obtained by assuming a raindrop distribution (such as the Marshall-Palmer distribution) and a constant collision[8] efficiency.

Sportisse [44] reviews such parametrizations of scavenging of radionuclides, focussing on below-cloud scavenging since data on in-cloud scavenging are sparse. Taking into account the spread in the estimation of the representative raindrop diameter, the scavenging coefficient (Λ in s^{-1}) for a monodisperse aerosol distribution is given by

$$\Lambda \simeq [0.43 - 1.71] \times 10^{-3}\, E\, r^{[0.75-0.842]},$$

where E is the collision efficiency and r is the rain intensity in mm hr^{-1}. For different particle sizes, E ranges between $[0.1 - 1.0]$ which leads to

$$\Lambda \simeq \left[4.3 \times 10^{-5} - 1.7 \times 10^{-3}\right] r^{[0.75-0.842]}.$$

Mircea and Stefan [26] show that the scavenging coefficient, integrated over the raindrop distribution, can be put into the form $\Lambda = Ar^B$ assuming a Gamma distribution function for the raindrop distribution and a constant collision efficiency. This leads to

$$\Lambda \simeq \begin{cases} [0.753 - 0.875]\, E\, r^{[0.78-0.86]}, & \text{for strong rains (100 - 500 mm hr}^{-1}\text{)}, \\ [1.91 - 1.95]\, E\, r^{[0.78-0.86]}, & \text{for moderate rains (5 - 100 mm hr}^{-1}\text{)}, \\ [20.56 - 26.67]\, E\, r^{[0.78-0.86]}, & \text{for weak rains (1 - 5 mm hr}^{-1}\text{)}, \end{cases}$$

where here the scavenging coefficient Λ is in hr^{-1} and E is a constant. With $E \simeq 0.1$ and after a

[8] The terms 'collision efficiency' and 'collection efficiency' are both used within the literature.

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Reference	Scavenging parameters (A;B) or scavenging coefficients Λ (in s^{-1})			
	Default	Particulate	Elemental Iodine	Organic Iodine
Raes et al. [31]	$(5 \times 10^{-5}; 0.8)$			
Päsler-Sauer [29]		$(8 \times 10^{-5}; 0.8)$	$(8 \times 10^{-5}; 0.6)$	$(8 \times 10^{-7}; 0.6)$
Belot et al. [5] & Caput et al. [7]	8.2×10^{-5} (gas)	$(4 \times 10^{-5}; 1)$, if $r_p < 1$ $(1.3 \times 10^{-4}; 1)$, if $r_p > 1$	$(4 \times 10^{-5}; 0.6)$	
CEC [8]		5×10^{-5}, if $r_p < 1$ 10^{-4}, if $r_p = 5$ 5×10^{-4}, if $r_p = 10$	10^{-4}	10^{-6}

Table 7: Scavenging parameters (A; B) from Sportisse [44] (scavenging coefficient $\Lambda = Ar^B$ in s^{-1}). For constant scavenging coefficients only one value (Λ) is given. The particle radius, r_p, is in μm.

conversion to s^{-1}, one obtains $\Lambda \simeq 2.3 \times 10^{-5}\, r^{0.8}$ for heavy rains and $\Lambda \simeq 6.6 \times 10^{-4}\, r^{0.8}$ for weak rains. A similar approach is used in Andronache [1] in order to justify the parametrization $\Lambda = Ar^B$ for a Marshall-Palmer distribution of raindrops and a range of particle sizes. Empirical models derived from measurements after the Chernobyl accident suggest $\Lambda \simeq \left[10^{-5} - 10^{-3}\right] r^{[0.5-0.7]}$ for caesium and iodine, with an average value of $\Lambda = 10^{-4}\, r^{0.64}$ [20]. Extensive measurements of ultrafine particles over forests in southern Finland gave scavenging coefficients ranging from 7×10^{-6} to 4×10^{-5} s^{-1} [24]. Table 7 shows other scavenging parametrizations taken from Sportisse [44].

Little is known about scavenging by snow and, furthermore, there is a lot of scatter in the experimental data. Jylhä [21] estimates a weak scavenging coefficient (Λ below 10^{-6} s^{-1}) for wet deposition of sulphur by snow. Sparmacher et al. [43], however, show that below-cloud scavenging coefficients for snow are about five times larger than washout coefficients for rain. Mircea and Stefan [26] present scavenging coefficients for solid precipitation of differing types and element shapes, varying precipitation intensities and for a range of assumed precipitation element distribution functions. The given scavenging coefficients have significant variation,

$$\Lambda = [8.8 - 117.8]\, E r^{[0.89-1.14]},$$

where Λ is in hr^{-1} and E is the collision efficiency which, in itself, has exhibited a wide spread of values from 0.01 to 1.0 [26].

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B Parametrizations of wet deposition in other models

We summarise here wet deposition schemes in other models. Note it is not always clear in the literature or model documentation whether the schemes described are used in-cloud and / or below-cloud, with rain and / or snow, with convective and / or dynamic (large-scale) precipitation, etc. Indeed some models do not make a distinction.

B.1 ADMS

In ADMS, wet deposition is modelled using the concept of a scavenging coefficient, Λ. (Note that the term 'washout coefficient' is used in the ADMS documentation. The model documentation states, however, that no distinction is made between in-cloud and below-cloud scavenging so, to avoid any confusion here, we use the generic term 'scavenging coefficient'.) The scavenging coefficient can be input directly by the user or estimated from other data such as rainfall rate. No distinction is made between different types of rain (frontal, convective, orographic, etc.). If Λ is specified as a constant value by the user then it is independent of the precipitation rate and, indeed, wet deposition will be predicted in the absence of precipitation. The default value for the scavenging coefficient, Λ, is 1.0×10^{-4} s^{-1} which, although typical of scavenging coefficients used elsewhere, could be considered large given its liberal application. Otherwise the scavenging coefficient is calculated from

$$\Lambda = Ar^B,$$

where r is the rainfall rate in mm hr^{-1} and A and B are parameters depending on the type of pollutant. The constants A and B are given by the user and take default values of $A = 1.0 \times 10^{-4}$ and $B = 0.64$.

For SO_2 and CO_2 the uptake is limited by the raindrop pH. The scheme calculates the aqueous concentration in the rainfall without considering the limiting effect of pH. This concentration (M_1) is then compared with the maximum aqueous concentration (M_2) that would be permitted by considering the pH of the raindrop. If $M_2 < M_1$, Λ is replaced by $\Lambda M_2/M_1$, thereby limiting the wet deposition.

A more complex option now exists for modelling wet deposition in ADMS and which allows reversibility of the uptake of pollutants by precipitation elements. This 'falling drop' method can be used for wet deposition modelling of SO_2 and HCl.

B.2 FLEXPART

The Lagrangian model FLEXPART takes into account a number of removal processes including wet deposition [46]. In-cloud and below-cloud scavenging are now treated differently within FLEXPART. The occurrence of clouds is calculated based on humidity and temperature information from the input meteorological data. If relative humidity is greater than 80% then cloud is assumed.

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B.2.1 In-cloud scavenging

Different in-cloud scavenging schemes are used for gases and aerosols and are based on the scheme of Hertel et al. [16]. The scavenging coefficient (Λ in s^{-1}) depends on the precipitation rate (r in mm hr^{-1}) and the height over which scavenging takes place (H_i, the cloud depth in m), and is obtained from

$$\Lambda = \frac{S_i\, r}{3.6 \times 10^6\, H_i},$$

where S_i is the in-cloud scavenging ratio which differs for gases and particles. The parametrization by Hertel et al. [16] is intended for convective conditions but is used in FLEXPART with both large-scale and convective precipitation.

For particles S_i is given by $S_i = 0.9/cl$ where cl is the cloud liquid water content (in m^3 water per m^3 cloud) calculated using

$$cl = 2 \times 10^{-7}\, r^{0.36}.$$

Hence

$$\Lambda = \frac{1.25}{H_i}\, r^{0.64}.$$

For gases S_i is given by $S_i = 1/cl_{eff}$ where cl_{eff} is an effective cloud liquid water content given by

$$cl_{eff} = \frac{(1 - cl)}{H_{eff} RT} + cl,$$

where H_{eff} is Henry's law constant for gases (in M atm^{-1}), R is the universal gas constant ($R = 8.2057 \times 10^{-2}$ atm M^{-1} K^{-1}) and T is the temperature of the gas (in K).

B.2.2 Below-cloud scavenging

The scheme for below-cloud scavenging uses a typical scavenging coefficient, Λ, given by

$$\Lambda = A\, r^B, \tag{9}$$

where r is the rainfall rate in mm hr^{-1} and Λ is in s^{-1}. The below-cloud scavenging parameters (A and B) for different species are given in table 8. FLEXPART uses the same scavenging parameters for snow and rain.

Since the below-cloud wet deposition parametrization depends nonlinearly on precipitation, account needs to be taken of subgrid variability of precipitation [16]. The area fraction F which experiences precipitation, for a given precipitation rate, is calculated by

$$F = \max\left[0.05, cc\frac{r_l\, fr_l(r_l) + r_c\, fr_c(r_c)}{r_l + r_c}\right],$$

where cc is the total cloud cover, r_l and r_c are the large-scale and convective precipitation rates in mm hr^{-1}, respectively and fr_l and fr_c are correction factors that depend on r_l and r_c (see table 9).

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Species	A	B
NO	8.0×10^{-6}	0.62
NO_2	1.0×10^{-5}	0.62
HNO_3	5.0×10^{-5}	0.62
H_2O_2	1.0×10^{-4}	0.62
NH_3	9.9×10^{-5}	0.62
SO_4	5.0×10^{-6}	0.62
NO_3	5.0×10^{-6}	0.62
I_2-131	8.0×10^{-5}	0.62
I-131	1.0×10^{-4}	0.80
Cs-137	1.0×10^{-4}	0.80
Y-91	1.0×10^{-4}	0.80
Ru-106	1.0×10^{-4}	0.80
Sr-90	1.0×10^{-4}	0.80
Wet tracer	1.0×10^{-4}	0.62
PCB28	5.0×10^{-6}	0.62
PCB101	5.0×10^{-6}	0.62
PCB180	5.0×10^{-6}	0.62
TEST	1.0×10^{-2}	0.62

Table 8: FLEXPART below-cloud scavenging parameters for different species

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The subgrid scale precipitation rate, r_s, is then given by

$$r_s = \frac{r_l + r_c}{F},$$

and is used in equation (9) as the rainfall rate. The depletion is applied to a fraction F of the mass.

Factor	$r \leq 1$	$1 < r \leq 3$	$3 < r \leq 8$	$8 < r \leq 20$	$20 < r$
fr_l	0.50	0.65	0.80	0.90	0.95
fr_c	0.40	0.55	0.70	0.80	0.90

Table 9: Correction factors used for the calculation of the area fraction that experiences precipitation. Precipitation rates, r, are in mm hr^{-1}.

B.3 CAMx

In CAMx (Comprehensive Air quality Model with eXtension) [13] scavenging of gases and aerosols in-cloud and below-cloud is based on a scavenging coefficient and uses relationships described by Seinfeld and Pandis [37]. Input to the CAMx wet deposition scheme includes three-dimensional cloud and precipitation water contents as liquid, snow (stratiform precipitation) and ice / graupel (convective precipitation) components.

For gases, the aqueous-phase scavenging coefficient (Λ_a) is given by

$$\Lambda_a = \Lambda_c \frac{c_{aq} L_c}{c \rho_w},$$

where c_{aq} is the aqueous concentration (mass per volume of water), L_c is the cloud liquid water content (in g m^{-3}), ρ_w is the liquid water density (in g m^{-3}) and c is the total concentration given by $c = c_g + c_{aq} L_c / \rho_w$, where c_g is the gas concentration (mass per volume of air). Λ_c is the scavenging coefficient for cloud water given by

$$\Lambda_c = 4.2 \times 10^{-7} \frac{Er}{d_d},$$

where E is the collection efficiency (0.9), r is the precipitation rate (in mm hr^{-1}) and d_d is the raindrop diameter (in m).

The gas-phase scavenging coefficient (Λ_g) is given by

$$\Lambda_g = 1.67 \times 10^{-6} \frac{r K_c}{d_d v_d},$$

where v_d is the fall speed of the raindrop and K_c is a mass transfer coefficient for a falling hydrometeor with fall speed v_d and diameter d_d,

$$K_c = \frac{D_g}{d_d} \left[2 + 0.6 \left(\frac{v_d d_d}{\nu} \right)^{1/2} \left(\frac{\nu}{D_g} \right)^{1/3} \right],$$

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where D_g is the molecular diffusivity of the gas and ν is the molecular diffusivity of the air.

In-cloud, the gas-phase and aqueous-phase scavenging coefficients are added to provide the total in-cloud scavenging rate for gases,

$$\Lambda = \Lambda_g + \Lambda_a.$$

Below-cloud the scavenging rate is given by the gas-phase scavenging coefficient,

$$\Lambda = \Lambda_g.$$

A limit due to the solubility of the gas and according to Henry's law is enforced.

For aerosols, an assumption that, within cloud, all aerosols exist within cloud water is made. Consequently, the scavenging coefficient for cloud water is used in-cloud ($\Lambda = \Lambda_c$). Below-cloud the scavenging coefficient for cloud droplets (Λ_c) is adopted but, in this case, the collection efficiency (E) is a function of the particle size, d_p.

B.4 ISCST3

In the Gaussian plume model ISCST3 (the Industrial Source Complex Short Term model), the scavenging coefficient (Λ in s^{-1}) is computed from a parameter (λ in (s mm hr^{-1})$^{-1}$) and the precipitation rate (r in mm hr^{-1}) [34],

$$\Lambda = \lambda r.$$

The parameter λ depends on the characteristics of the pollutant (e.g., solubility and reactivity of gases and the particle size distribution for aerosols) as well as the nature of the precipitation (e.g., liquid or frozen). The parameter λ is input by the user and the user's attention is drawn to best-fit values reported as a function of particle size by Jindal and Heinold [18]. The work of Jindal and Heinold [18] suggest that λ takes a minimum value of about 4 $\times$ 10^{-5} hr s^{-1} mm^{-1} for particles with diameters approximately 0.6 to 1 μm and a maximum value of about 6.6 $\times$ 10^{-4} hr s^{-1} mm^{-1} for particles approximately 10 μm and larger in diameter. The coefficients used with frozen precipitation are expected to be about 1/3 of the values suggested by Jindal and Heinold [18].

B.5 CALPUFF

The scavenging coefficient (Λ in s^{-1}) in the Gaussian puff model CALPUFF [35] is calculated using

$$\Lambda = \lambda \left(\frac{r}{r_1} \right),$$

where λ is a scavenging parameter (in s^{-1}), r is the precipitation rate (in mm hr^{-1}) and r_1 is a reference precipitation rate of 1 mm hr^{-1}. The parameter λ depends on the characteristics of the

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pollutant (e.g., solubility and reactivity) as well as the nature of the precipitation and the default values for SO_2, SO_4^{2-}, NO_x, HNO_3 and NO_3^- are given in table 10.

Pollutant	CALPUFF scavenging parameter, λ (s^{-1})	
	Liquid precipitation	Frozen precipitation
SO_2	3×10^{-5}	0
SO_4^{2-}	1×10^{-4}	3×10^{-5}
NO_x	0	0
HNO_3	6×10^{-5}	0
NO_3^-	1×10^{-4}	3×10^{-5}

Table 10: Default scavenging coefficients for a precipitation rate of 1 mm hr^{-1} (λ in s^{-1}) in CALPUFF

B.6 EMEP

The wet deposition scheme in the Unified EMEP model includes both in-cloud and below-cloud scavenging and distinguishes between gases and particles [6, 39]. There is currently no distinction between liquid precipitation (rain) and frozen precipitation (snow and ice) scavenging.

B.6.1 In-cloud

In-cloud scavenging is calculated using

$$\Delta C_{wet} = -C \frac{W_{in} P}{\Delta z \rho_w},$$

where ΔC_{wet} is the wet deposition per second, C is the concentration, W_{in} is the in-cloud scavenging ratio, P is the precipitation rate (in kg m^{-2} s^{-1}) obtained by summing convective and stratiform precipitation, Δz is the characteristic scavenging depth (assumed to be 1000 m) and ρ_w is the water density (1000 kg m^{-3}). Converting the precipitation rate into mm hr^{-1} (r) gives

$$\Delta C_{wet} = -C \frac{W_{in}}{3.6 \times 10^9} r,$$

with a scavenging coefficient (in s^{-1}), $\Lambda = W_{in}\, r/(3.6 \times 10^9)$. Assumed values for W_{in} for different pollutants are given in table 11.

B.6.2 Below-cloud

Below-cloud scavenging of gases is calculated using

$$\Delta C_{wet} = -C \frac{W_{sub} P}{\Delta z \rho_w} = -C \frac{W_{sub} r}{3.6 \times 10^9},$$

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Pollutant	W_{in} $(\times 10^6)$	$\frac{W_{in}}{3.6 \times 10^9}$ $(\times 10^{-4})$	W_{sub} $(\times 10^6)$	$\frac{W_{sub}}{3.6 \times 10^9}$ $(\times 10^{-4})$	$\bar{E}$	$\frac{A\,\bar{E}}{3.6 \times 10^3\,V_{dr}}$ $(\times 10^{-4})$
SO_2	0.3	0.83	0.15	0.42		
HNO_3	1.4	3.9	0.5	1.4		
NH_3	1.4	3.9	0.5	1.4		
H_2O_2	1.4	3.9	0.5	1.4		
HCHO	0.1	0.28	0.03	0.083		
SO_4^{2-}	1.0	2.8			0.1	0.29
NO_3^- (fine)	1.0	2.8			0.1	0.29
NH_4^+	1.0	2.8			0.1	0.29
$PM_{2.5}$	1.0	2.8			0.1	0.29
NO_3^- (coarse)	1.0	2.8			0.4	1.2
PM (coarse)	1.0	2.8			0.4	1.2

Table 11: Wet scavenging ratios and collection efficiencies used in the Unified EMEP model

where W_{sub} is the sub-cloud scavenging ratio given in table 11 for different species.

Below-cloud scavenging of particles is based on Scott [36] and calculated using

$$\Delta C_{wet} = -C\,\frac{A\,P}{V_{dr}}\,\bar{E} = -C\,\frac{A\,\bar{E}}{3.6 \times 10^3\,V_{dr}}r,$$

where V_{dr} is the raindrop fall speed (V_{dr} = 5 m s^{-1}), A = 5.2 m^3 kg^{-1} s^{-1} is an empirical coefficient (a Marshall-Palmer size distribution is assumed for raindrops) and $\bar{E}$ is the size dependent collection efficiency of aerosols by raindrops given in table 11.

B.7 The DERMA model

The wet deposition scheme for aerosols in the Danish Emergency Response Model of the Atmosphere (DERMA) [2, 3, 40, 41, 42] is based on the concept of a scavenging coefficient and is strongly influenced by the original wet deposition scheme in NAME. The scavenging coefficient represents a washout coefficient for below-cloud scavenging and a rainout coefficient for in-cloud scavenging. Below-cloud the collection efficiency, and hence the washout coefficient, have been shown to be strongly dependent on the particle size (the so-called 'Greenfield gap'). Näslund and Holmström [28] express the below-cloud washout coefficient (Λ in s^{-1}) as a polynomial function of particle radius (r_p) and rainfall rates (r in mm hr^{-1}),

$$\Lambda(r_p, r) = \begin{cases} a_0\,r^{0.79}, & r_p < 1.4\,\mu m, \\ \left(b_0 + b_1\,r_p + b_2\,r_p^2 + b_3\,r_p^3\right) f(r), & 1.4\,\mu m < r_p < 10\,\mu m, \\ f(r), & r_p > 10\,\mu m, \end{cases} \tag{10}$$

where $f(r) = a_1\, r + a_2\, r^2$, $a_0 = 8.4 \times 10^{-5}$, $a_1 = 2.7 \times 10^{-4}$, $a_2 = -3.618 \times 10^{-6}$, $b_0 = -0.1483$, $b_1 = 0.3320133$, $b_2 = -3.0062 \times 10^{-2}$ and $b_3 = 9.34458 \times 10^{-4}$. To improve the washout parametrization by including the mechanism of Brownian capture, Baklanov and Sørensen [3] present an additional formulation of the washout coefficient,

$$\Lambda = \frac{r}{2a_m}\left[\frac{4}{Pe}\left(1 + 0.4Re^{1/2}Sc^{1/3}\right) + \frac{4r_p}{a_m}\left(\frac{r_p}{a_m} + \frac{(1 + 2\mu_w r_p/\mu_a a_m)}{\left(1 + Re^{-1/2}\mu_w/\mu_a\right)}\right) + \left(\frac{\rho_w}{\rho_a}\right)^{1/2}\left(\frac{St - St^*}{St - St^* + 2/3}\right)^{3/2}\right],$$

where $Re = a_m w_r/\mu$ is the Reynolds number, $St = \left(-2r_p^2\rho_p w_r/9a_m\rho_a\nu\right)$ is the Stokes number, $St^* = \left[1.2 + \ln\left(1 + Re/2\right)/12\right]/\left[1 + \ln\left(1 + Re/2\right)\right]$ is the critical Stokes number, $Sc = \nu/D$ is the Schmidt number, $Pe = a_m w_r/D$ is the Peclet number, ν is the kinematic viscosity, D is the Brownian diffusivity of particles, μ_w and μ_a are the dynamic viscosity of water and air, ρ_p and ρ_a are the density of particles and air, a_m is the volume mean raindrop projected radius (taken to be $a_m = 0.35(r/r_0)^{-1/4}$ mm for steady frontal rain), w_r is the mean vertical velocity of the raindrops, r is the rain rate (in mm hr^{-1}), r_0 is a nominal rainfall rate of 1 mm hr^{-1} and r_p is the particle radius (in μm). There is no distinction made between below convective cloud scavenging coefficients and below dynamic cloud scavenging coefficients.

In-cloud rainout by convective precipitation is more effective than washout. The original NAME parametrization (equation (7)) is suggested for in-cloud rainout by convective precipitation. For dynamic precipitation, the below-cloud washout coefficient given by equation (10) is recommended for in-cloud rainout. The presence of clouds is determined from relative humidity fields.

Processes of scavenging by snow are not well understood and hence the parametrizations used in the original NAME scheme (equations (4) and (5)) are recommended with no dependence of particle radius.

B.8 The Unified Model: the CLASSIC aerosol scheme

The wet deposition parametrization in the Coupled Large-scale Aerosol Simulator for Studies in Climate (CLASSIC) aerosol scheme in the Unified Model (MetUM) treats scavenging by large-scale and convective precipitation separately and differently [19]. For convective precipitation the assumption that cloud only occupies 5% of the grid-box area is made. In addition rainout (or in-cloud) scavenging is treated differently to washout (or below-cloud) scavenging. Wet deposition is calculated within each model layer.

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B.8.1 Sulphate aerosols

Rainout of dissolved sulphate by large-scale precipitation from within a model layer is calculated by

$$\Delta \{SO_4\} = \{SO_4\} \left[\exp\left(-f_{diss}\,\beta\, t\right) - 1\right]$$

where $\{SO_4\}$ is the mixing ratio of dissolved sulphate before the rainout, f_{diss} is the fraction of dissolved sulphate that is dissolved in the cloud droplet (f_{diss} = 1), β is the conversion rate of condensed water into precipitated water (in s^{-1}) and t is the time-step (in s).

Rainout of dissolved sulphate by convective precipitation is parametrized using

$$\frac{\mathrm{d}C}{\mathrm{d}t} = -\lambda_c\, r\, C,$$

where $\lambda_c = 3 \times 10^{-5}$ s^{-1} and r is the total precipitation rate (in mm hr^{-1}) including snow.

Washout of sulphate aerosols in particle form by all types of precipitation (convective and large-scale) is neglected since it is considered less important than rainout.

B.8.2 Sulphur dioxide

The washout parametrization of SO_2 by both large-scale and convective precipitation reflects both the solubility of SO_2 and the possibility of raindrops becoming saturated. It is based on a scavenging coefficient (Λ in s^{-1}) which depends on the concentration by volume (S in ppbv) of SO_2 and the rain rate (r in mm hr^{-1}). Scavenging by snow is neglected. The scavenging coefficient is given by

$$\Lambda(r,S) = \begin{cases} \lambda_0\, r^{2/3}, & \text{for } S \le S_0, \\ \lambda_1 \left(\dfrac{r}{S}\right)^{2/3} & \text{for } S > S_0, \end{cases}$$

where $\lambda_0 = 6.5 \times 10^{-5}$ s^{-1}, $\lambda_1 \simeq 2.955 \times 10^{-5}$ s^{-1} and $S_0 = 0.3065$ ppbv. As above, convective precipitation is assumed to take place over just 5% of the horizontal area and hence the precipitation rate within convective shower regions is obtained by dividing the grid-box precipitation rate by 0.05.

There is no specific rainout parametrization of sulphur dioxide in the UM. Once dissolved in cloud droplets, gases undergo aqueous phase chemistry that converts mass into the aerosol phase. The washout parametrization is also applied, however, at model levels within cloud where there is rain (i.e., it implicitly includes some rainout).

B.8.3 Ammonia

The washout parametrization of NH_3 by both large-scale and convective precipitation is based on the assumption that ammonia is sufficiently soluble that raindrops can never become saturated. The

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scavenging coefficient, Λ, is therefore independent of the concentration and is calculated by

$$\Lambda = \lambda_a \, r^{2/3},$$

where Λ is in s^{-1}, r is the rain rate (in mm hr^{-1}) and $\lambda_a = 1.0 \times 10^{-4}$ s^{-1}.

In the same way as sulphur dioxide, there is no specific rainout parametrization of ammonia but the washout parametrization also represents some scavenging from within cloud.

B.8.4 Dimethyl sulphate

Wet scavenging of DMS is not represented in the model.

B.8.5 Black carbon / soot

Washout of soot by large-scale precipitation is currently turned off in the model since rainout dominates wet deposition of soot by large-scale precipitation. The rainout parametrization by large-scale precipitation is calculated in the same way as described for sulphate aerosols in section B.8.1.

For convective precipitation, washout of soot is neglected and rainout of soot is parametrized using a scavenging coefficient, $\Lambda = 0.108\,P$, where Λ is in s^{-1} and P is the total convective precipitation rate (rain and snow) in kg m^{-2} s^{-1}. Converting the precipitation rate into mm hr^{-1} gives $\Lambda = 3 \times 10^{-5} r$, where r is the convective precipitation rate in mm hr^{-1}.

B.8.6 Biomass smoke aerosol

Washout of biomass aerosol by large-scale and convective precipitation is neglected. Rainout of biomass aerosol by large-scale and convective precipitation is calculated in the same way as for soot as described in section B.8.5.

B.8.7 Organic carbon aerosol derived from fossil-fuel combustion

Wet scavenging of organic carbon aerosols derived from fossil-fuel combustion (OCFF) is treated in the same way as wet scavenging of biomass-burning aerosols (see section B.8.6).

B.8.8 Nitrate aerosol

Ammonium nitrate, in either particle mode or dissolved mode, is subject to scavenging by convective precipitation. Dissolved nitrate is also subject to rainout, but not washout, by large-scale precipitation. Wet scavenging of nitrate is treated in the same way as the particle mode and dissolved mode sulphate (see section B.8.1).

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B.8.9 Dust aerosols

Dust particles are not hydrophilic so rainout of dust aerosols is relatively unimportant and is neglected. Washout by convective and large-scale precipitation is calculated using a scavenging coefficient, $\Lambda = K\,r$, where the coefficient K depends on the particle size and r is the rainfall rate in mm hr^{-1}. Table 12 gives the values of K for six particle size bins. The K values are derived from the experimental data of Volken and Schumann [47].

bin	radius (μm)	K
1	$0.0316 - 0.1$	2.0×10^{-5}
2	$0.1 - 0.316$	2.0×10^{-5}
3	$0.316 - 1.0$	3.0×10^{-5}
4	$1.0 - 3.16$	6.0×10^{-5}
5	$3.16 - 10.0$	4.0×10^{-4}
6	$10.0 - 31.6$	4.0×10^{-4}

Table 12: Values of the coefficient K for six dust particle size bins.

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